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RAE structure and satellite lines in the photon-excited *K* X-ray spectrum of fluorine

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Abstract : In the photon-excited *K* X-ray spectrum of fluorine, the RAE structure (emission of photon along with an electron in addition to an electron filling the *K*-vacancy) and K_α satellite lines have been observed, using plane crystal spectrometer. The $K_\alpha L^n$ satellite line energies and relative intensities have been measured with respect to the K_α diagram line. The RAE structure in fluorine caused by photon-excitation is being reported for the first time to the best of our knowledge.

Keywords : Photon-excitation, *K* X-ray spectrum, satellite lines

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1. Introduction

Experimental measurements using crystal spectrometers provide an opportunity to look for atomic transitions where single *K*-plus-*L*-shell ionization as well as multiple electron-vacancy rearrangement occurs with one or more inner-shell vacancies. The satellite lines [1–3] form the group of lines corresponding to the transitions from initial states having one hole in the *K*-shell and *n* holes in the *L*-shell, represented by $K_\alpha L^n$ $(1s)^{-1}$ $(2p)^{-n} \rightarrow (2p)^{-n-1}$. The normal diagram line corresponds to $n = 0$ *i.e.* $K_\alpha L^0$, a vacancy transfer $(1s)^{-1} \rightarrow (2p)^{-1}$. The study of satellite lines is of vital importance to provide information about the multiple *L*-shell ionization and hence on intra-atomic electron correlations, excitation dynamics and other effects influencing the *X*-ray emission process.

The other type of transitions correspond to the process in which more than one vacancy undergoes rearrangement and a single photon is emitted from an atom with a

K-shell vacancy. These single-photon two-electron rearrangement processes are called radiative Auger effect (RAE) and radiative electron rearrangement (RER). In the RAE process [4–9], the decay of a *K*-shell vacancy proceeds as a normal *K* Auger process except that there is an emission of photon along with an electron in addition to an electron filling the *K*-vacancy while in the RER process [9,10], a *K* X-ray is emitted coincident with two 2*s* electrons undergoing rearrangement such that one of the 2*s* electron fills the *K*-shell vacancy while the other is promoted to the 2*p* subshell. The RAE produces a broad structure, in the *K* X-ray spectra, with energy less than the $K_{\alpha_{1,2}}$ diagram line and has been clearly visible in proton bombardment [7], heavy ion bombardment [9], electron excitation [8] and ionization caused by photons [4,8]. The RAE edge, corresponding to high energy side of the structure, corresponds to zero velocity of the ejected electron and the photon having approximately the same energy as a $KL_{23}L_{23}$ Auger electron [9]. Further, the RAE has been found to be independent of the mode of excitation as observed by Richard *et al* [7] and Sirvola *et al* [8].

The measurements of satellite lines in fluorine have been extensively carried out by ion impact [11,12] However, only one paper by Tachibana *et al* [13] has appeared on K_α satellite lines by photon excitation using NaF and KF samples. These authors [13] have experimentally observed that the intensity of the first satellite peak with respect to the diagram line of KF was much reduced than that of NaF although the theoretical values of the transition probability for both these cases are the same. They further concluded that the resonant electron transfer model is unable to explain such a dramatic change. Using electron bombardment, the relative intensities of the satellite lines with respect to the diagram line *i.e.* $K_\alpha L^1 / K_\alpha L^0$ and $K_\alpha L^2 / K_\alpha L^0$ for various elements $9 \leqq Z \leqq 20$ were measured by Fischer [14]. On the theoretical side, the first X-ray satellite intensities of neon-like ions in the sudden approximation, have been calculated by Åberg [15] for $9 \leqq Z \leqq 20$ and indicate a systematic decrease in the relative intensities with increasing Z. The average energy shifts of the K_α satellite lines with respect to the diagram lines for atoms in the range of $8 \leqq Z \leqq 20$ have been calculated by Natarajan [16] using relativistic screened hydrogenic wave functions and have been measured for a few element in the range $11 \leqq Z \leqq 19$ by Murthy *et al* [17] using photon excitation.

The present measurements are aimed to find the RAE and satellite lines in fluorine using LiF sample using photon bombardment to look for the multiple ionization caused by photons, because (i) there is no experimental data on the low energy side of the diagram line in fluorine using photon excitation; (ii) the first satellite line ($K_\alpha L^1$) of fluorine observed by 84 MeV N^{4+}-ions [11] and 5.5 MeV He^+-ions [12] is found to have large variation in intensity depending not only on the chemical environment, but also differ by about a factor of 2 in these two experiments; (iii) to check whether the intensity of the first satellite line with respect to the diagram line differs from the observations made by Tachibana *et al* [13] using photon bombardment on NaF and KF samples.

2. Experimental procedure and data analysis

The X-ray diffractometer (RIGAKU Model 9579) consisted of Machlett Rh anode 3 kW X-ray tube, a plane crystal spectrometer with Thallium Acid Phosphate (TAP) (100) crystal and a continuous Ar + 10%CH_4 gas flow (50 ml/min) proportional counter. The LiF single crystal sample was housed in an evacuated chamber. Two fine collimators (0.01°) were used one each at the target and the detector. The X-ray photons scattered in a given direction are reflected by the analyzing crystal. The X-ray tube was operated at 50 kV and 50 mA. The whole system was housed in a room with constant temperature. The energy calibration of the system was done using K_α X-ray lines from standard target materials like Al and Mg. The K X-ray spectrum (RAE and satellite) of fluorine was recorded in first order using TAP crystal ($2d$ = 25.763 Å). The spectra were recorded in Bragg angle (2θ) steps of 0.05° for 200 seconds at each angle over the desired angular region. For the background measurements, 30 to 40 pieces of filter papers were piled up in the place of actual target materials and the spectrum was recorded again as described above. The angle of the wave dispersive data was transformed into the corresponding energy scale to display as in Figure 1. The spectrum was analysed in terms of least square peak fitting procedure. For

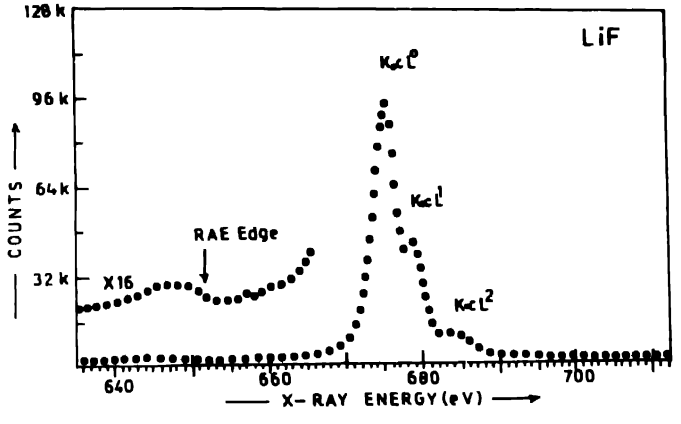

Figure 1. The photon-induced K X-ray spectrum of LiF recorded with the TAP crystal in the first order The spectrum clearly indicates the two satellite lines ($K_\alpha L^1$ and $K_\alpha L^2$) along with the $K_\alpha L^0$ diagram line of fluorine. The broad peak structure due to the Radiative Auger Effect (RAE) and the RAE edge are shown on the expanded counts-scale

this purpose, an interactive computer code NFIT [18] workable on personal computer was developed. The fitting method involves the minimization of the residue between the raw data and the fitted components and is based on the Gauss-Newton method, which is faster than the usual simplex method. This code can fit 15 peaks with three parameters (peak-height, -position and -FWHM) for each peak along with a linear background. The choice can be made to fix some of the parameters to make residue minimum. The code makes the appropriate initial values. However, being interactive, each parameter can be inputted according to the desired initial value, if necessary.

3. Results and discussion

The presently measured energies and relative intensities of satellite transitions together with that of the diagram line, are given in Table 1. The table also lists the theoretical and measured values by other workers. The errors shown in the present results for various transitions arise due to the (i) dispersal of the scattered angles of the fluorescent X-rays by

Table 1. Energy and Intensity values of K X-ray diagram line and satellite lines of fluorine measured by a plane crystal spectrometer The energy corresponding to the RAE edge is also given

Transition	Energy (eV)		Relative intensity	
	Present measurements	Other workers	Present measurements	Other workers
RAE edge	649 ± 1			
$K_\alpha L^0$ ($K_{\alpha_{1,2}}$)	675 3 ± 0.1	676 8 [19]	100	
		677 0 [16]		
$K_\alpha L^1$ satellite	679 4 ± 0.1	681 6 [16]	38 5 ± 2 5	42 to 108 [11]
				39 to 102 [12]
				39 5 ± 1 5 [14]
				42.5 [15]
$K_\alpha L^2$ Satellite	684 6 ± 0.2		10.5 ± 0 7	32 to 55 [11]
				18 to 31 [12]

the optical system; (ii) imperfections of the crystal (tilts between small grains formed in the preparation); (iii) inaccuracy of the lattice constant of the crystal; and (iv) temperature dependence of the lattice constant. The errors in the intensity values for different transitions arise from (i) the counting statistics, (ii) the variation of the efficiency of the proportional counter with energy values of different transitions, (iii) selection of the background under the peaks, and (iv) the fitting procedure for the peaks.

The energy of the diagram line has been found to be 1.5 eV lower than those of the Bearden and Thomsen [19] value while the present energy values for the diagram and the first satellite line are 1.7 eV lower than the theoretically values predicted by Natarajan [16]. The intensity of the first satellite line of fluorine has been found to be (38.5 ± 2.5)% as compared to the diagram line, which is in fair agreement with the value 39.5% for NaF measured by electron excitation [14] and theoretically predicted value (42.5%) of Åberg [15]. The first and second satellite line intensities for almost all the compounds of fluorine (NaF, Na_3AlF_6, AlF_3, NiF_2 and CuF_2) reported by Uda *et al* [11], due to 84 MeV N^{4+}-ion bombardment and the results of Benka *et al* [12] due to 5.5 MeV He^+-ion bombardment on CsF, RbF, BaF_2, KF, SrF_2, CaF_2 and NaF show a large variation in the values for different compounds of fluorine. The present value of $K_\alpha L^1 / K_\alpha L^0$ in fluorine, using LiF sample, agrees well within experimental error to the lower limit of approximately 42% for NaF obtained by Uda *et al* [11] and for SrF_2 (39%) and KF (40%) obtained by Benka *et al* [12]

in their range of values as shown in Table 1. Tachibana *et al* [13] tried to explain their observed difference in the photon-induced first K_α satellite intensity in fluorine for NaF and KF samples using a reconstruction scheme of molecular orbitals with numerical basis SCC-DV-X_α method in the ionized or hole states. This approach predicts the same transition probability (≈ 88%) in these two compounds of fluorine. No comparison with the present results using LiF sample could however, be made due to their unquoted experimental values of satellite intensity in the case of these two fluoride samples. The second satellite line intensity $K_\alpha L^2 / K_\alpha L^0$ measured in the present case is much smaller than the value quoted for any of the fluorine samples employed by Uda *et al* [11] and Benka *et al* [12].

The separation energies of the first and second satellite from the diagram line are 4.1 ± 0.2 and 9.3 ± 0.3 eV respectively while the separation energy between the first satellite line and the diagram line predicted by Natarajan [16] is 4.6 eV. The average of configuration value of the energy shift with zero *M*-shell electron as calculated by Maurer and Watson [20] for multiple *K*-plus-*L*-shell vacancy states of fluorine is only 2.9 eV which is too low in comparison to 4.1 ± 0.2 eV.

The present measurements indicate that the multiple ionization by photons causes the RAE structure and emission of $K_\alpha L^n$ satellite transitions along with the diagram line. This indicates that the photons produce multiple *L*-vacancies along with *K*-vacancy like heavy charged particles. However, the intensity of the diagram line, in each case, is always greater than the satellite lines contrary to the heavy ion bombardment. Since the ionization mechanism in the photon bombardment is different from that of charged particle bombardment [13], a theoretical model is needed to explain the results of photon bombardment in a fairly satisfactory manner.

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